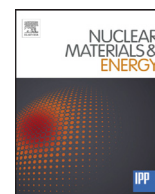


Contents lists available at [ScienceDirect](http://ScienceDirect)

## Nuclear Materials and Energy

journal homepage: [www.elsevier.com/locate/nme](http://www.elsevier.com/locate/nme)

# Fluence dependent changes of erosion yields and surface morphology of the iron-tungsten model system: SDTrimSP-2D simulation studies

U. von Toussaint<sup>a,\*</sup>, A. Mutzke<sup>b</sup><sup>a</sup>Max-Planck-Institute for Plasmaphysics, Boltzmannstrasse 2, Garching D-85748, Germany<sup>b</sup>Max-Planck-Institute for Plasmaphysics, Wendelsteinstrasse 1, Greifswald D-17491, Germany

## ARTICLE INFO

## Article history:

Received 21 August 2016

Revised 31 August 2016

Accepted 12 September 2016

Available online xxx

## Keywords:

Fusion

Plasma-facing materials

Sputter yield

First wall

Monte Carlo simulations

Surface enrichment

TRIM

## ABSTRACT

The effect of different sample structures of an iron-tungsten model system (as a surrogate for reduced activation ferritic martensitic steels like EUROFER) on the development of surface morphologies, tungsten surface enrichment and sputter yields under low-energy monoenergetic perpendicular 200 eV deuterium bombardment has been studied with SDTrimSP-2d simulations. Previous modeling studies considering diffusive effects also could reasonably reproduce and explain the experimental results for a large set of experimental parameters like temperature, flux and sample concentration. However, for settings with negligible Fe-W-interdiffusion the fluence needed for steady-state conditions differed between the experiments and the simulations. Thus, the main focus of the present study is directed towards the elucidation of this fluence mismatch. Comparison of one and two-dimensional simulation results reveal a strong dependency of the tungsten enrichment on the sample homogeneity and a significantly delayed reduction of the erosion yield due to a pronounced formation of surface structures from initially flat sample surfaces.

© 2016 The Authors. Published by Elsevier Ltd.

This is an open access article under the CC BY license (<http://creativecommons.org/licenses/by/4.0/>).

## 1. Introduction

Usage of bare reduced activation ferritic martensitic (RAFM) steel tiles has recently been considered as a possible option for the plasma-facing component in the far-SOL region of a future fusion reactor. RAFM steels like EUROFER contain important concentrations of heavy elements with tungsten being the most prominent component. These different components in RAFM steels will be eroded differently, leading to changes in surface composition and erosion yield, where the sputter yield of eg. EUROFER decreases to less than 10% of the iron sputter yield at very high fluences [1]. Ion beam analysis showed an enrichment of W at the exposed surface [2] correlating with the yield reduction and binary-collision based simulations including solid-state diffusion on iron-tungsten model systems agreed qualitatively with the measured data for different sample compositions and temperatures [3]. Unfortunately, the limited experimental depth resolution did not allow to confirm the predicted W enrichment within the first monolayers unambiguously. Therefore, it still remains unclear whether the W enrichment is the only mechanism underlying the yield reduction or if also topography changes contribute to the observed effects as recent SEM

studies seem to indicate. However, without detailed investigation of the counteracting effects of roughening surfaces on the erosion, i.e. an increase of the sputter yield by non-perpendicular impact as well as effective surface area and a decrease by the reduced particle escape probability even the sign of the effect is hard to predict, not even to mention the magnitude or the fluence dependence. Here we present a detailed study of the 2D-surface morphology evolution and the sputter yields of the iron-tungsten model system under 200 eV deuterium impact for different samples.

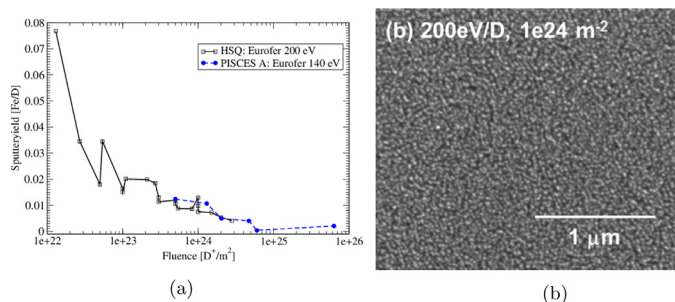
## 2. Experimental observations

In several previous studies [1,2,4,5] sputtering of iron-tungsten model systems and RAFM steels like EUROFER and F82H under low-energy deuterium bombardment have been investigated. Although a wealth of experimental data is now available we focus subsequently on two aspects: a) the fluence dependence of erosion and b) morphology changes.

Generally a reduction of the sputter yield on fluence scales of the order of  $\sim 10^{25} \text{D/m}^2$  as indicated in Fig. 1a is observed. Subsequent analysis of exposed Fe-W layers by Rutherford backscattering backs the predicted enrichment process of W at the surface, although the surface layer cannot be resolved fully. However, an increasing smoothing of the iron-edge in the RBS spec-

\* Corresponding author.

E-mail address: [udo.v.toussaint@ipp.mpg.de](mailto:udo.v.toussaint@ipp.mpg.de) (U. von Toussaint).



**Fig. 1.** Left panel: Fluence dependence of the iron sputtering yield of EUROFER (dashed line) under exposure to a D plasma with a bias of -150 V (corresponding to a D energy of 140 eV) PISCES A. During exposure the sample temperature was in the range of 350 K–460 K [1]. Also shown is the sputter yield of EUROFER exposed to a monoenergetic beam of D (solid line). Right panel: SEM image from the surface of the Fe-W model system after irradiation with  $10^{24} D/m^2$  200 eV/D.

tra indicates the presence of roughening effects [2] under D bombardment. An SEM image of the Fe-W model system with an initial W-concentration of 0.7 at.% after exposure to  $10^{24} D/m^2$  is displayed in Fig. 1b (figure adapted from [2]). Although the image appears on a large scale pretty homogeneous, the formation of small structures not present in the as-deposited case (cf. [2], Fig. 2a) can already be recognized. This interpretation is also supported by SEM images of RAFM steels after exposure [4], which indicate an even stronger structure formation under D bombardment compared to the Fe-W system. At elevated temperatures (above  $\sim 900$  K) surface segregation of tungsten may also become important [5].

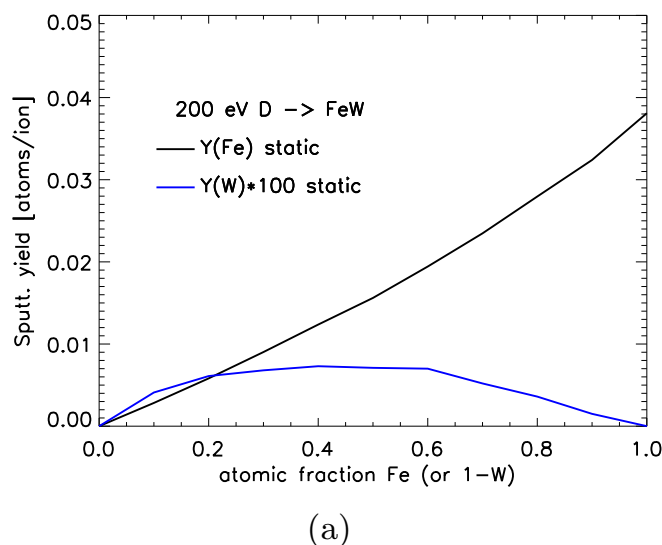
### 3. Computational approach

Many properties of the Fe-W layer system under low energy D-bombardment (200 eV) have recently been modelled by dynamic SDTrimSP-simulations taking into account also solid-state diffusion [3], i.e. the flux dependency of the W surface enrichment, the influence of the initial (homogeneous) tungsten concentration and of exposure temperature. The simulation results based on a 1-D sample structure (i.e. the sample composition is only a function of depth  $\tilde{c} = \tilde{c}(x)$ ) show generally good agreement with mea-

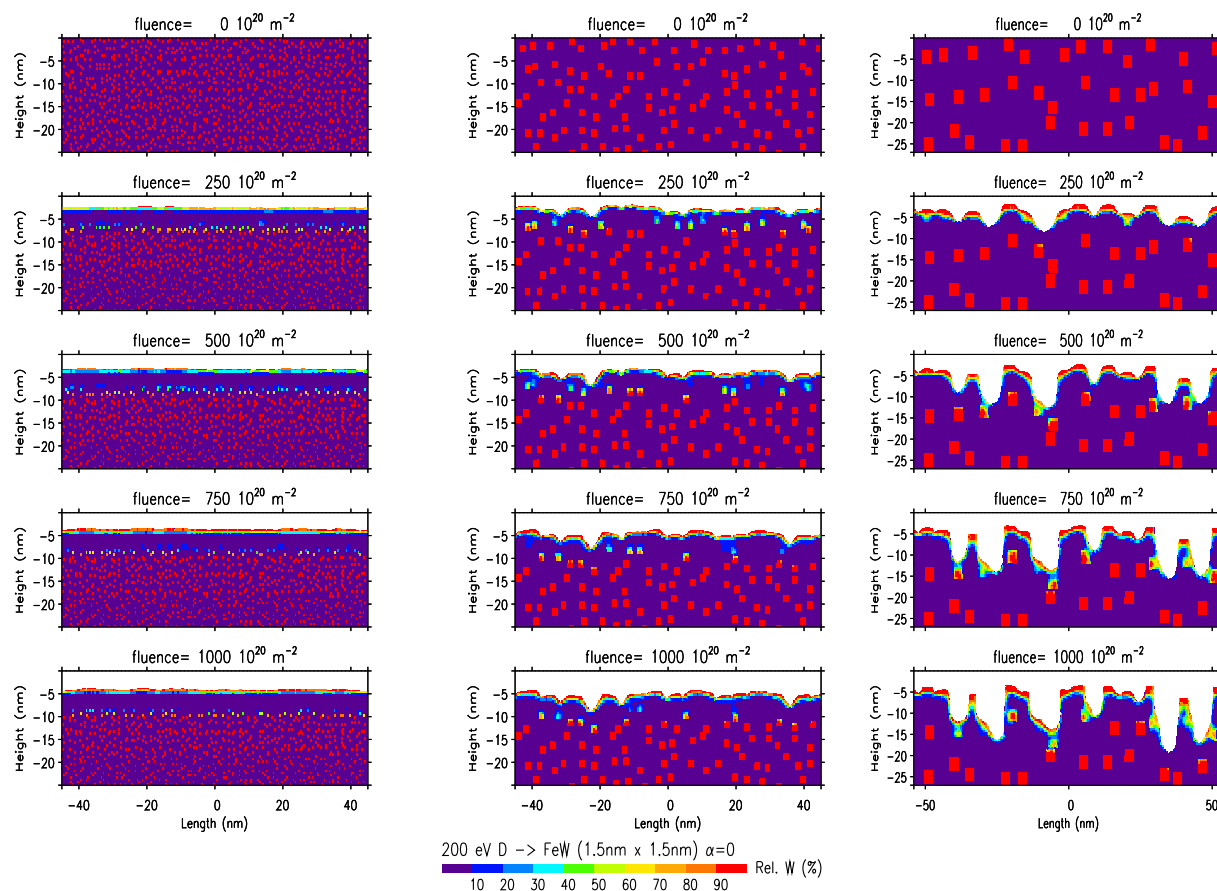
surements. Many of the observed effects can be rationalized by a non-monotonic sputter yield dependency of tungsten on the sample composition. As can be seen from Fig. 2a where data derived from a 1D SDTrimSP simulation are displayed, the sputter yield of iron increases monotonically—as it is to be expected—with the iron concentration in the sample. In contrast, the sputter yield of tungsten is zero for a pure tungsten sample but exhibits a broad maximum for tungsten concentrations in the range of 20% to 60% W. The bombarding species is monoenergetic deuterium with an energy of 200 eV and perpendicular impact. For this deuterium energy the energy transfer to a tungsten atom is below the sputter threshold, such that tungsten sputtering can only occur via energy transfer by an intermediate iron-tungsten collision. This explains the non-monotonic variation of the tungsten sputter-yield as function of the iron concentration: if there is no iron present, tungsten cannot be sputtered.

The biggest discrepancy between the measured data and the simulation results is the observation of a much faster convergence of the erosion yield towards the steady-state value with D fluence in the simulations compared to the measurements. As can be seen from Fig. 1a convergence to steady-state requires typically fluences of at least  $O(10^{24} D/m^2)$  to  $O(10^{25} D/m^2)$ . In contrast, the simulated iron depth distribution is rapidly approaching the steady-state profile. In Fig. 2b depth profiles for the sequence of 0, 1, 50, 100, 150, 200, 250 and  $300 \times 10^{20} D/m^2$  of a sample with an initially constant concentration profile of 90at.% iron and 10at.% tungsten are displayed. The deuterium energy was 200 eV, the deuterium flux  $10^{15} D/m^2 s$  and the sample temperature was set to 573 K (thus negligible diffusion). Already at  $O(10^{22} D/m^2)$  the profiles do hardly change and beyond a fluence of  $O(10^{22} D/m^2)$  the sputter yield has become constant. The increase of the iron concentration above the bulk sample concentration of 90at.% Fe is due to the continuous forward implantation of iron recoils into the subsurface region.

There are many possible explanations for this difference between experiment and simulation, as eg. small amounts of heavy plasma impurities, which suppress tungsten enrichment. However, given that it is a quite general observation in several devices and together with the information provided by the SEM images, the questionable (although ubiquitous) assumption of a smooth surface in the simulations could also be responsible for the difference. To



**Fig. 2.** Left panel: Sputter yield of tungsten and iron for 200 eV deuterium (perpendicular impact) as function of the atomic fraction of iron. The unusual non-monotonic behavior of the tungsten sputter yield is due to the required intermediate deuterium-iron collision for tungsten sputtering. Right panel: Iron distributions as function of depth for different 200 eV deuterium fluences. The sample temperature was 573 K and a deuterium flux of  $10^{15} D/m^2 s$  was applied. The initial sample composition was 90at.% iron and 10at.% tungsten.



**Fig. 3.** Evolution of the surface morphology and concentration profiles under perpendicular bombardment with 200 eV D ions for different sample structures of the same overall composition. The figures in the left hand column display the change in composition (color-coded) and surface structure as function of deuterium fluence (the fluence increases from top to bottom) of an iron matrix with embedded tungsten particles of size  $5 \text{ Å} \times 5 \text{ Å}$ . The middle column and the right column display the same fluence series but with embedded tungsten particles of size  $15 \text{ Å} \times 15 \text{ Å}$  and  $30 \text{ Å} \times 30 \text{ Å}$ , respectively.

clarify this we need to model the consequences of surface roughness and thus inhomogeneous samples.

### 3.1. SDTrimSP-2D

The physical model implemented in the Monte Carlo code family SDTrimSP for the simulation of ion-solid interactions is based on the binary collision approximation [6]. The present model allows to calculate ion-transport in amorphous materials (since the positions of the collisions partners are chosen randomly from an appropriate distribution), the formation of collision cascades in three dimensions and induced mixing processes.

However, it was also noted that the angular dependence of the sputtering yield severely hampers approaches to describe surfaces with a pronounced roughness by 1D models [7,8], although attempts have been made to consider the effect of roughness by weighted averaging of surface patches at inclined angles [9] or by a fractal model of the surface [10]. For that reason the SDTrimSP-2D code (for a description of the code see [11]) has been developed to simulate the interaction of impinging energetic particles with 2D non-planar surfaces. It has been validated in a series of experiments using several target systems, including tungsten surfaces and optical gratings [12–14].

## 4. Computation and results

The simulations are computationally demanding due to the need of small fluence steps and the additional spatial dimension which needs to be sampled. To avoid excessive running times

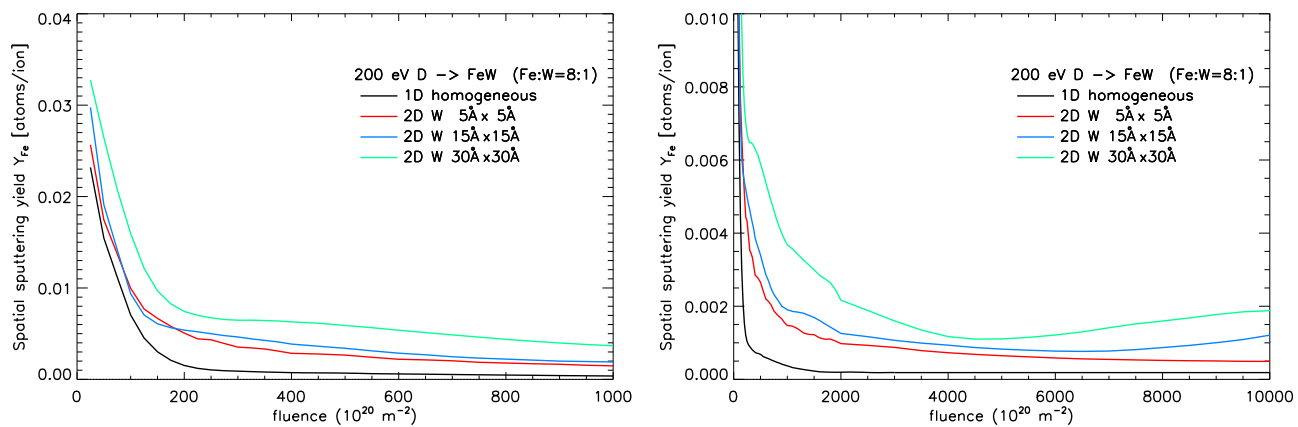
solid-state diffusion was not considered and the results apply to conditions where solid-state diffusion can be neglected, i.e. sample temperatures below  $\sim 600 \text{ K}$  [3]. Collision-cascade induced mixing effects are taken into account, however, more subtle effects like enhanced (surface) diffusion by ion-bombardment or ion-induced segregation which may become relatively more important at lower temperatures are not included in the present model.

### 4.1. Sample composition

The average sample composition used in the study had a iron-tungsten ratio of 8:1 by volume which, considering the respective atomic densities, converts into an atomic fraction of 91.5% iron and 8.5% tungsten. However, the sample structure has been varied: to take into account the inhomogeneous distribution of tungsten in RAFM-steels as well as the agglomeration of tungsten observed in scanning electron microscopy images [2] of the iron-tungsten model system the available tungsten has been spread as elemental pure tungsten blocks throughout a pure iron matrix. The size of the tungsten blocks has been varied, from  $5 \text{ Å} \times 5 \text{ Å}$  up to  $30 \text{ Å} \times 30 \text{ Å}$ , where the simulation lattice parallel and perpendicular to the surface was matched to the smallest length scale of  $5 \text{ Å}$ .

### 4.2. Results

Fig. 3 displays the evolution of the sample morphology as function of deuterium fluence from top to bottom. The left panel has tungsten blocks of size  $5 \text{ Å} \times 5 \text{ Å}$  embedded in the iron matrix. Already at deuterium fluences of  $250 \times 10^{20} \text{ D/m}^2$  a thin ( $\sim 1 \text{ nm}$ )



**Fig. 4.** The left hand panel displays the fluence dependence of iron sputtering yields of Fe-W systems under perpendicular bombardment with 200 eV D ions for different sample structures of the same composition up to  $1 \times 10^{23} \text{D/m}^2$ . The right hand panel displays also the iron sputtering yields of the Fe-W systems but now extended to a deuterium fluence of  $10 \times 10^{23} \text{D/m}^2$ . Please note the magnified y-axis.

but clearly recognizable surface layer with increased tungsten concentration has been formed. Underneath that layer an apparent depletion of tungsten takes place, which becomes even more pronounced with increasing fluence. However, this ‘depletion’ is in fact an increase of the iron density due to recoil (forward) implantation of iron into the subsurface layers. This effect of recoil implantation can also be seen in the concentration profiles given in Fig. 2b. Above a fluence of  $1000 \times 10^{20} \text{D/m}^2$  the profile is essentially constant and the surface stays flat and smooth. This is also reflected in the fluence dependence of the sputter yield, displayed in the left panel of Fig. 4. Within the first  $200 \times 10^{20} \text{D/m}^2$  there is a rapid reduction of the sputter yield from  $Y = 0.03 \text{ atoms/D}$  by a factor of three. Thereafter the decrease slows down considerably and saturates around  $Y = 5 \times 10^{-4} \text{ atoms/D}$  for  $1 \times 10^{24} \text{D/m}^2$ .

In the middle panel tungsten is randomly distributed in blocks of size  $15 \text{ Å} \times 15 \text{ Å}$ . With increasing fluence the surface exhibits some structure and becomes rough on a scale of several nanometers. Nevertheless, although delayed with respect to the former case at a fluence of  $1000 \times 10^{20} \text{D/m}^2$  an almost closed tungsten surface layer has been formed. Up to this fluence the sputter yield dependency is not too different from the previous one. However, a small difference remains even up to the largest simulated fluences of  $1 \times 10^{24} \text{D/m}^2$ .

The right panel of Fig. 3 reveals a strong built-up of surface morphology with increasing fluence. This surface structure consists of pins which are covered by tungsten layers and gaps with significant lower surface concentrations of tungsten. The protective W surface coverage eventually results in almost perpendicular side walls. The large scale of the formed structures (a consequence of the  $30 \text{ Å} \times 30 \text{ Å}$  tungsten blocks) and the related surface dynamics did not allow to reach a steady-state condition. Rough estimates based on the changes of the surface morphology point to fluences well above  $1 \times 10^{25} \text{D/m}^2$ . These large scales can also be seen in the right panel of Fig. 4, where at fluences of  $1 \times 10^{24} \text{D/m}^2$  the sputter yield still shows an increasing trend, indicating that the surface is far from equilibrium.

It should be noted that simulations with an impact angle of 30 degrees result in almost identical surface morphologies developments and sputter yield dependencies.

## 5. Conclusion and outlook

Simulations of Fe-W models systems under low-energy (200 eV) deuterium bombardment were performed using SDTrimSP-simulations (1D-simulations for perfectly smooth sur-

faces and homogeneous sample compositions) and 2D-simulations for inhomogeneous samples. In the case of inhomogeneous samples the structure size of tungsten ‘precipitates’ has decisive influence on the sputter yield and on the fluence dependent development of the sputter yield. Larger tungsten particles result in initially larger sputter yields and in increased fluence scales until steady-state conditions are obtained. These increased fluence scales exceed the fluences required for convergence in the 1D case by orders of magnitude and thus resemble much better the fluence scales needed in experiments to reach steady-state. Therefore the consideration of the surface morphology development in the Fe-W system appears to be crucial.

Of special interest in the ongoing studies is the effect of small amounts of heavy impurities on the tungsten surface enrichment, the morphology and the associated sputter yields. Not only will in future fusion devices seeding gases be used for radiative cooling and thus also interact with the walls but also because in most present day plasma devices impurities cannot be fully avoided and therefore their consideration may be crucial for the interpretation of the experimental results.

## References

- [1] J. Roth, K. Sugiyama, V. Alimov, T. Höschel, M. Baldwin, R. Doerner, EUROFER As wall material: Reduced sputtering yields due to w surface enrichment, *J. Nucl. Mater.* 454 (2014) 1–6.
- [2] K. Sugiyama, Erosion study of fe-W binary mixed layer prepared as model system for RAFM steel, *J. Nucl. Mater.* 463 (2015) 272–275.
- [3] U. von Toussaint, A. Mutzke, K. Sugiyama, T. Schwarz-Selinger, Simulation of coupled sputter-diffusion effects, *Physica Scripta* T167 (2016) 014023. <http://stacks.iop.org/1402-4896/2016/i=T167/a=014023>.
- [4] V. Alimov, Y. Hatano, K. Sugiyama, M. Balden, T. Höschel, M. Oyaidzu, J. Roth, J. Dörner, M. Fußeder, T. Yamanishi, Surface modification and deuterium retention in reduced activation ferritic martensitic steels exposed to low-energy, high-flux d plasma and d<sub>2</sub> gas, *Physica Scripta* T159 (2014) 014049.
- [5] M. Balden, S. Elgeti, M. Zibrov, K. Bystrov, T.W. Morgan, Temperature effect on the surface morphology, d retention and erosion of EUROFER exposed to low-energy high-flux d plasma, *Nucl. Mater. Energy* submitted (2016) xxx.
- [6] W. Möller, B.M.U. Scherzer, Modeling of hydrogen implantation into graphite, *J. Appl. Phys.* 64 (1988) 4860.
- [7] M. Küstner, W. Eckstein, E. Hecht, J. Roth, Angular dependence of the sputtering yield of rough beryllium surfaces, *J. Nucl. Mater.* 265 (1–2) (1999) 22–27.
- [8] I. Bizyukov, K. Krieger, N. Azarenkov, U. von Toussaint, Relevance of surface roughness to tungsten sputtering and carbon implantation, *J. Appl. Phys.* 100 (2006) 113302–1–113302–7.
- [9] M. Küstner, W. Eckstein, V. Dose, J. Roth, The influence of surface roughness on the angular dependence of the sputter yield, *Nucl. Instrum. Meth. B* 145 (1998) 320–331.
- [10] D.N. Ruzic, The effect of surface roughness characterized by fractal geometry on sputtering, *Nucl. Instrum. Methods Phys. Res. Section B* 47 (1990) 118–125.

- [11] A. Mutzke, R. Schneider, G. Bandelow, SDTrimSP-2D: Simulation of Particles Bombarding on a Two Dimensional Target - Version 2.0, Technical Report IPP 12/11, Max-Planck-Institut für Plasmaphysik, Garching, 2013.
- [12] A. Mutzke, R. Schneider, I. Bizyukov, SDTrimSP-2D studies of the influence of mutual flux arrangement on erosion and deposition, J. Nucl. Mater. 390–391 (2009) 115–118, doi:[10.1016/j.jnucmat.2009.01.133](https://doi.org/10.1016/j.jnucmat.2009.01.133).
- [13] I. Bizyukov, A. Mutzke, R. Schneider, J. Davis, Evolution of the 2D surface structure of a silicon pitch grating under argon ion bombardement: Experiment and modeling, Nucl. Instrum. Methods Phys. Res. Section B 268 (17–18) (2010) 2631–2638, doi:[10.1016/j.nimb.2010.06.035](https://doi.org/10.1016/j.nimb.2010.06.035).
- [14] A. Mutzke, I. Bizyukov, H. Langhuth, M. Mayer, K. Krieger, R. Schneider, Study of interaction of c+ ion beam with a si pitch grating on a macro-scale level, Nucl. Instrum. Methods Phys. Res. B 293 (2012) 11–15, doi:[10.1016/j.nimb.2012.09.017](https://doi.org/10.1016/j.nimb.2012.09.017).